

Anion Formation in Sputter Ion Sources by Neutral Resonant Ionization

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A widespread theory of surface ionization explains negative secondary ionization mass spectrometry (SIMS) and is extended to Cs sputter ion sources (CSIS), but SIMS and CSIS differ greatly in analytical practice. Primary ion beams sputter analyzed surfaces in SIMS at controlled erosion rates while CSIS uses a primary ion beam with 10^3 to 10^5 higher power densities to rapidly erode a material. Highly focused Cs^+ beams in CSIS sources create mm-diameter sputter craters that support small blue plasma balls on which negative ion currents and ionization efficiencies are notably dependent. Sputtering produces overwhelmingly neutral products that the plasma has long been thought to ionize as in a charge-change vapor, but taking place at 1000-fold lower energies. Neutral atoms at keV energy were shown to capture electrons from neutral Cs in the 1960's and calculations showed in the 1970's that cross sections rose dramatically at lower energies with Cs atoms in excited states. Resonant behavior was demonstrated in the 1980's with neutral resonant ionization (NRI) occurring at high rates for eV energy collisions.

A collision-radiation model of a Cs plasma in a recess was developed that followed electronic excitation and optical resonance up to the Cs(7d) state to understand control of the excited states. NRI cross sections are essentially unknown, but the Landau-Zener-Stückelberg formalism predicts that the maxima rise as the inverse square of the energy deficit: the difference between the ionization potential of the Cs state and the electron affinity of the sputtered neutral atom. The model shows production of Cs excited states under conditions that maximize measured CSIS ion current, explaining much "lore" of sputter sorcery. The model has been directly tested. The predicted reduction of $^{36}\text{S}^-$ ions by addition of Ir or Pt to AgCl samples for ^{36}Cl AMS is seen from suppression of Cs(5d). The high level of Cs(7d) in a 0.5 mm recess explains the $83 \mu\text{A}/\text{mm}^2$ C^- current density compared to the $20 \mu\text{A}/\text{mm}^2$ from a 1 mm recess that is a strong refutation of the surface ionization hypothesis of CSIS operation.